Magnetic ordering of Pr in $Pb_2Sr_2PrCu_3O_8$

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The magnetic ordering of the Pr ions in $Pb_2Sr_2PrCu_3O_8$ has been studied using neutron-diffraction and ac-suspectibility measurements. An imperfect three-dimensional magnetic ordering of the Pr spins was observed at a temperature well below its Neel temperature of $T_N \approx 7$ K. The magnetic intensities observed at $T = 1.4$ K can be explained by assuming long-range order in the ab plane with short-range correlations, of correlation length $\xi_c = 20$ Å, along the c axis and a moment directed along the c-axis direction. The magnetic ordering is 2D in nature, and the basic magnetic structure consists of nearest-neighbor spins that are aligned antiparallel along all three crystallographic directions. The magnetic transition is also evident in the ac-susceptibility versus temperature measurements, where a cusp that is typical of antiferromagnetic ordering is clearly observed, which matches the T_N obtained by neutron diffraction.

Just like other high-T_c oxides, $Pb_2Sr_2PrCu_3O_8$ (Pr 2:2:1:3) also possesses a layered structure. From the crystallographic structural point of view, the major difference between Pr 2:2:1:3 and $PrBa_2Cu_3O_7$ (Pr 1:2:3) is the number of layers stacked along the c-axis direction. The structure of the former can be obtained by replacing the CuO-chain layer in the latter with two PbO layers and one Cu layer.[1] This replacement results in even more anisotropic physical properties for the 2:2:1:3 systems than for the 1:2:3 systems. The rare-earth atoms in the 2:2:1:3 systems form an orthorhombic sublattice, where $a \approx b$ and the nearest-neighbor distance along the c axis is more than four times that in the ab plane. It is thus clear that the crystallographic anisotropy naturally leads to highly anisotropic magnetic interactions, and two-dimensional (2D) behavior can be expected.

The 3D long-range ordering of the Pr spins has been observed in both the Pr 1:2:3 and $TlBa_2PrCu_2O_7$ (Pr 1:2:1:2) systems with anomalously high ordering temperatures.[2,3] Numerous observations have indicated that hybridization plays an important role in the Pr magnetism. If this is the case we should then expect the effect along the c-axis direction to be much reduced in the Pr 2:2:1:3 system compared to the Pr 1:2:3 and Pr 1:2:1:2 systems, simply because of its wider spacing along the c axis. The Pr 2:2:1:3 system is then a better candidate for observing the 2D character of the Pr ordering. In this paper we report an imperfect 3D ordering of the Pr spins observed in Pr 2:2:1:3. The Pr spins ordered at 7 K, nevertheless even at $T = 1.4$ K, the correlations between the Pr spins along the c-axis direction are still short range. The basic magnetic structure consists of Pr spins that are aligned antiparallel along all three crystallographic directions. The antiferromagnetic ordering of the Pr spins is also evident in the ac-susceptibility versus temperature measurements, where a cusp, typical of antiferromagnetic ordering, in the χ_{ac} vs T curve is clearly evident.

A powder sample of $Pb_2Sr_2PrCu_3O_8$ was prepared by the standard solid-state reaction technique, and the details can be found elsewhere.[4] Both x-ray and high-resolution neutron diffraction were used to characterize the sample. The nearest-neighbor distances between the Pr atoms that we obtained at room temperature were 3.815(1) Å and 15.765(4) Å in the ab plane and along the c-axis direction, respectively. The nominal oxygen concentration determined from neutron profile refinement analysis was 8.01(2). During the course of the low-temperature neutron-diffraction experiment, the sample was sealed in a cylindrical holder filled with helium exchange gas to promote thermal conduction at low temperatures. A pumped ^{4}He cryostat was used to cool the sample, and the lowest temperature achieved was 1.4 K.

Neutron-diffraction measurements were performed at the Research Reactor at the U.S. National Institute of Standards and Technology. The data were collected using the BT-9 triple-axis spectrometer operated in double-axis mode. The incoming neutrons had a wavelength of 2.352 Å defined by a pyrolytic graphite PG(002) monochromator, with a PG filter placed after the monochromator position for suppressing higher-order wavelength contaminations. The angular collimations used were 40′ in front of the monochromator, and 48′-48′ before and after the sample position, respectively. No analyzer crystal was used in these measurements.

The magnetic signal was isolated from the nuclear one by subtracting the data collected at high temperatures from the data taken at low temperatures.[5] Figure 1 shows the magnetic Bragg peaks thus obtained at $T = 1.4$ K, where the diffraction pattern taken at $T = 15$ K, serving as the nonmagnetic "background," has been subtracted from the data. The indices shown are based on the Pr chemical unit cell. The underlying spin structure of Pr hence consists of the nearest-neighbor spins that are aligned antiparallel along all three crystallographic directions. This is the same type of spin structure found in Pr 1:2:3[2] and Pr 1:2:1:2.[3]

The widths of the observed magnetic peaks are much broader than the instrumental resolution, which indicates a short-range ordering of the Pr spins. Short-range correlations along the c-axis direction and long-range correlations within the ab plane can be expected, since the nearest-neighbor dis-
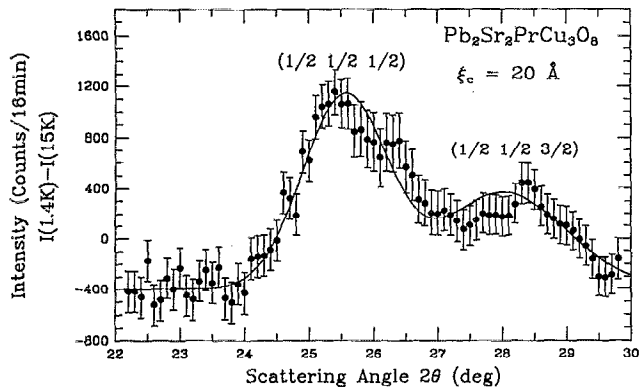
FIG. 1. Magnetic intensities observed in $Pb_2Sr_2PrCu_3O_8$ at $T=1.4$ K, where the indices shown are based on the Pr chemical unit cell. The Pr spins order antiferromagnetically along all three crystallographic directions. The solid curve is a fit to the data assuming long-range order in the ab plane and a short-range correlation length $\xi_c = 20$ Å along the c axis.
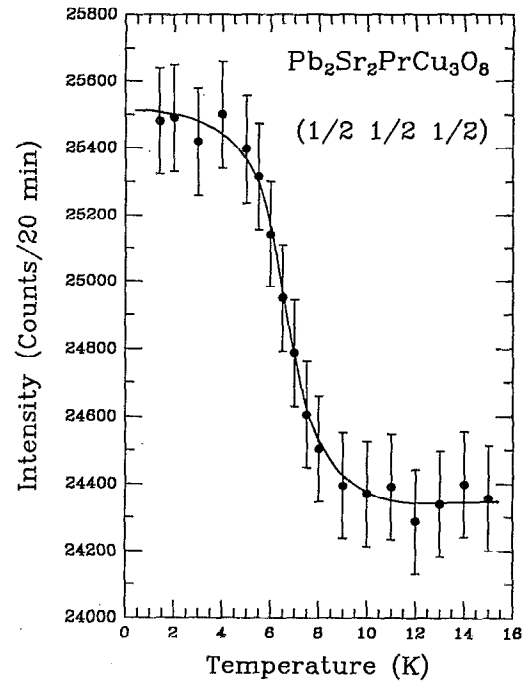

FIG. 2. Temperature dependence of the $\{\frac{1}{2}\frac{1}{2}\frac{1}{2}\}$ peak intensity. The data reveal a typical order parameter with an ordering temperature of $T_N \approx 7$ K. The solid curve is only a guide to the eye.

tance along the c axis is more than four times that in the ab plane. The solid line shown in Fig. 1 is a fit to the theoretical curve, assuming long-range correlations in the ab plane and short-range correlations, with a correlation length $\xi_c = 20$ Å, along the c axis and a moment pointed along the c-axis direction. In generating the fitted curve we adopt Warren's approach [6] of averaging the scattered intensity for a 2D powder sample over all possible orientations in reciprocal space, and in addition allow for the presence of finite correlations along the third axis.[7] In comparison with the Pr ordering in Pr 1:2:3 and Pr 1:2:1:2, the Pr-Pr coupling remains antiferromagnetic and the 2D character becomes more pronounced in Pr 2:2:1:3 likely due to the larger spacing between the Pr-Pr atoms along the c axis. This crystallographic character results in a reduction of the hybridization between the Pr ions that are mediated through the copper-oxygen layers between them.

Figure 2 shows the temperature dependence of the $\{\frac{1}{2}\frac{1}{2}\frac{1}{2}\}$ peak intensity, and reveals a typical order parameter with a Neel temperature of $T_N \approx 7$ K. This ordering temperature of the Pr spins is a factor of 2 smaller than that found in Pr 1:2:3:7 ($T_N \approx 17$ K) and is close to that found in Pr 1:2:1:2 ($T_N \approx 8$ K). The ordering temperature of Pr is still much too high for purely dipolar interactions to explain.

The ac-susceptibility measurements were performed using a Lake Shore 7221 ac susceptometer. Data were collected with an alternating magnetic field of strength 1 Oe and frequency 250 Hz. A portion of the real part of the measured $\chi_{ac}(T)$ is shown in Fig. 3(a), and the temperature dependence of its derivative $d\chi_{ac}/dT$ is shown in Fig. 3(b). The magnetic transition is clearly seen with a $T_N \approx 7$ K determined to be at the relative minimum in the $d\chi_{ac}/dT$ vs T plot. A cusp in the $\chi_{ac}-T$ curve, which is typical of antiferromagnetic ordering,[8] is also evident. These results are consistent with that obtained from neutron-diffraction measurements. We note that negative values for χ_{ac} were obtained on the high-temperature side shown in Fig. 3(a). This is due to the Meissner diamagnetism of the superconducting state present in the compound. The interplay between the Pr mag-

netism and superconductivity is the subject in a separate study.[9]

In conclusion, we have studied the magnetic ordering of the Pr spins in Pr 2:2:1:3 by neutron-diffraction and ac-susceptibility measurements. At $T=1.4$ K, which is well below the observed ordering temperature of $T_N \approx 7$ K, an im-
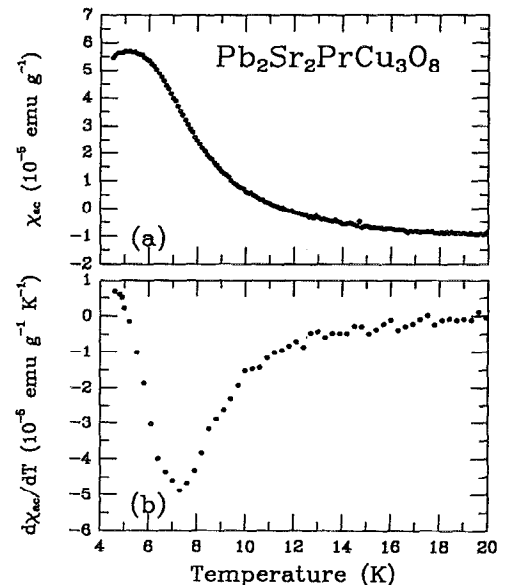

FIG. 3. A portion of the measured ac susceptibility $\chi_{ac}(T)$ and its temperature derivative $d\chi_{ac}/dT$ plotted against temperature. The transition temperature is determined to be at the relative minimum in the $d\chi_{ac}/dT$ vs T plot, which gives $T_N \approx 7$ K. This T_N is consistent with the ordering temperature obtained from the data shown in Fig. 2.

J. Appl. Phys., Vol. 76, No. 10, 15 November 1994

Hsieh et al.   7125

perfect 3D ordering of the Pr spins was observed, with long-range correlations in the *ab* plane and short-range correlations along the *c* axis. The 2D behavior is believed to arise naturally from the crystallographic structure, and results in a reduction of the hybridization between the Pr ions in the present system. The ordering temperature observed is still much too high for purely dipolar and RKKY interactions may be anticipated.

The Tb ions in the isostructural compound Tb 2:2:1:3 were found[10] to order at $T_N \approx 5.5$ K. Even at $T = 1.4$ K the ordering of the Tb spins is still purely 2D without any correlation found along the *c* axis. It is then clear that, in the 2:2:1:3 system the coupling between the Pr atoms is much stronger than between the Tb atoms along all three crystallographic directions. Moreover, short-range correlations between the Pr spins have also been observed[11] in the $PrBa_2Cu_3O_{6.2}$ compound, which suggests that the oxygen concentration may also have important influence on the Pr hybridization along the *c* axis. A study of the effects of the oxygen concentration on the Pr ordering in the 2:2:1:3 system should give further information on the nature of the Pr hybridization.

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